Carbon Ad-Dimer Defects in Carbon Nanotubes: A Route to Patterned Tubes for Energy Applications

M. Sternberg[a], L. Curtiss[a], D. Gruen[a], G. Kedziora[b], D. Horner[c], P. Redfern[a], and P. Zapol[a]
[a] Argonne National Laboratory; [b] HPTI, Wright Patterson AFB, OH; [c] North Central College, Naperville, IL

Motivation

Thermoelectric energy conversion is needed to increase energy supply. The key for thermoelectric technology is to find materials with the *figure of merit* $ZT>3$. This requires *independent control* of:

- Density of states at the Fermi level (increase S)
- Carrier concentrations (increase σ)
- Phonon scattering (reduce κ)

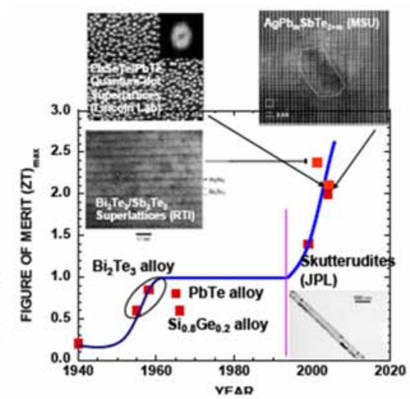
In bulk materials these properties are linked. By contrast, *nanoscale materials* may provide breakthroughs.

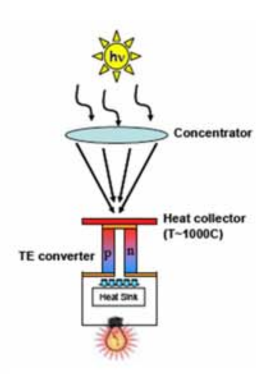
$$ZT = (S^2 \sigma / \kappa)\, T$$

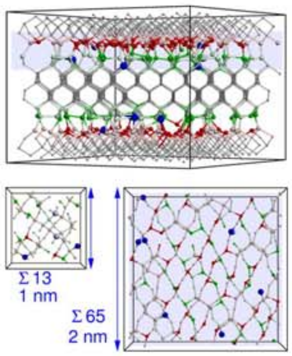
ZT	figure of merit
S	Seebeck coefficient
σ	electrical conductivity
κ	thermal conductivity

Figure: Progress timeline in thermoelectric materials. Source: BES Report *Basic Research Needs for Solar Energy Utilization* (2005).

Background





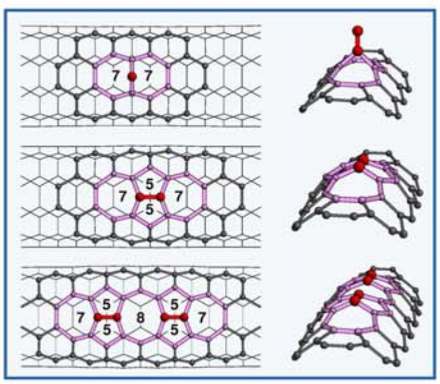


Novel Nanocarbon Materials

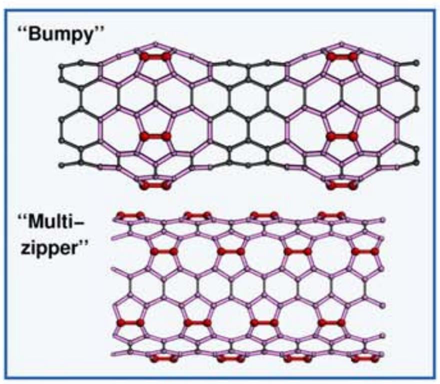
- Ultrananocrystalline diamond (UNCD)
  - 3–5 nm grain size.
  - Low thermal conductivity.
  - High electronic conductivity, but low carrier mobility.
- Patterned carbon nanotubes (CNT)
  - High electron mobility.
  - Reduced thermal conductivity.

Modeling results

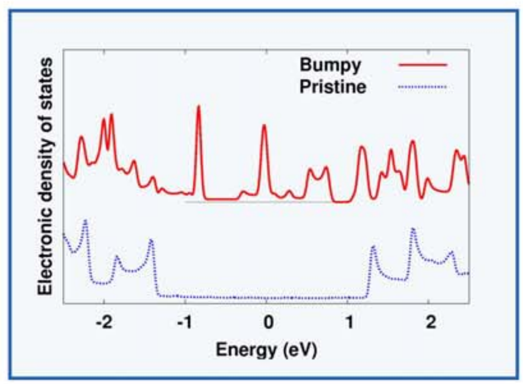
- Computer simulations predict stable structures of pristine and patterned carbon nanotubes.
- Carbon dimers (C_2) insert with low barriers into CNT walls (lower than for Stone-Wales defects).
- Adsorption energies and barriers depend on size and type of CNT.
- C_2 adsorption modifies the electronic structure of CNT - it introduces electronic states near the Fermi level.



- Stages for initial C_2 adsorption on a (5,5) nanotube, resulting in topological defects.



- Patterned tubes resulting from multiple controlled C_2 depositions.



- Electronic densities of states for a patterned tube. New peaks emerge near Fermi level.

Significance

- Opportunity for creating new device functionalities for a range of energy technology applications.
- Wide window for manipulation of peaks in the density of states near the Fermi level in the patterned CNTs.
- Low dimensional materials with densities of states similar to those predicted for the patterned nanotubes often display substantially improved thermoelectric performance and could provide efficient means for solar energy conversion.

Future Work

- Use theory and modeling to help guide synthesis of UNCD/CNT composites with covalent bonding and patterned nanotubes.
- Enhancement of thermoelectric performance.
- Apply modeling techniques for computer simulations of further new materials.

M. Sternberg, L. Curtiss, D. Gruen, G. Kedziora, D. Horner, P. Redfern, and P. Zapol, Phys. Rev. Lett., in print (2006)

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This work was supported by the U.S. Department of Energy, Basic Energy Sciences, under contract W-31-109-ENG-38.

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